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[Document Name] Patent Application [Case Number] 2037140059 [Date of Application] November 5, 2002 [Destination] Commissioner of the Japan Patent Office [International Patent Classification] H01M 8/00 [Inventor] [Address] c/o Matsushita Electric Industrial Co., Ltd. 1006, Oaza Kadoma, Kadoma shi, Osaka [Name] Aoi TANAKA [Inventor] [Address] c/o Matsushita Electric Industrial Co., Ltd. 1006, Oaza Kadoma, Kadoma-shi, Osaka [Name] Norihisa MINO [Patent Applicant] [Identification Number] 000005821 [Name] Matsushita Electric Industrial Co., Ltd. [Attorney] [Identification Number] 110000040 [Name] IKEUCHI-SATO & PARTNER PATENT ATTORNEYS [Representative] Hiroyuki IKEUCHI [Telephone Number] 06-6135-6051 [Official Fee] [Advance Payment Note Number] 139757 [Amount of Payment] 21000 [List of File Documents] [Name of Document] Specification 1 1 [Name of Document] Drawing

[Name of Document] Abstract

[Proof] Required

[General Power of Attorney's Number] 0108331

[Document Name] SPECIFICATION
[TITLE OF THE INVENTION] FUEL CELL
[CLAIMS]

[Claim 1] A fuel cell for generating an electric power by supplying one of electrodes with a fuel and the other with an oxidant,

wherein at least one of the electrodes comprises a thin film electrolyte, a catalyst and an electron-conducting substance,

the thin film electrolyte comprises an organic compound molecule that is chemically bonded to a surface of a substrate, and

the organic compound molecule comprises an ion-conducting functional group serving as an electrolyte.

[Claim 2] The fuel cell according to claim 1, wherein the chemical bond is at least one bond selected from the group consisting of a covalent bond, an ionic bond, a coordinate bond and a metallic bond.

[Claim 3] The fuel cell according to claim 1 or 2, wherein the chemical bond is a covalent bond formed by an elimination reaction.

[Claim 4] The fuel cell according to claims 1 to 3, wherein the chemical bond is a bond to the surface of the substrate via an oxygen atom.

[Claim 5] The fuel cell according to claim 1, wherein the ion-conducting functional group is at least one functional group selected from the group consisting of a phosphonyl group, a phosphinyl group, a sulfonyl group, a sulfonyl group, a sulfonyl group, a sulfonic group, a mercapto group, an ether linkage group, a hydroxyl group, a quaternary ammonium group, an amino group and a phosphate group, and comprises an ion.

[Claim 6] The fuel cell according to claim 1, wherein the substrate is a catalyst particle, an electron—conducting moving particle, or an inorganic compound or an organic compound that is further added to the electrode.

[DETAILED DESCRIPTION OF THE INVENTION]

[0001]

[Technical field to which the invention pertains]

The present invention relates to a fuel cell that uses directly hydrogen, methanol, ethanol, dimethyl ether, isopropyl alcohol, ethylene glycol, glycerin, methane, dimexymethane and the like as a fuel and the air, oxygen or ozone as an oxidant.

[0002]

[Prior Art]

A fuel cell generates electric power by an electrochemical reaction between a fuel capable of generating a hydrogen ion such as hydrogen and an oxygen containing oxidant such as the air. Its structure will be described herein. First, catalyst layers are formed respectively on both surfaces of a polymer electrolyte for transporting hydrogen ions selectively. Next, on outer surfaces of these catalyst layers, gas diffusion layers are formed using, for example, a water-repellent electrically conductive carbon particle paper that has both fuel gas permeability and electron conductivity. The catalyst layer and the gas diffusion layer form an electrode. [0003]

Then, a gas sealant or a gasket is disposed so as to surround the electrode and sandwich the polymer electrolyte so that a fuel to be supplied may not leak out and be mixed with the oxidant. This sealant or gasket is integrated with the electrode and the polymer electrolyte, thus forming a membrane electrode assembly (MEA).

[0004]

In general, the catalyst layer of the fuel cell is produced by preparing a paste of a platinum-based precious metal catalyst as a catalyst with electrically conductive carbon particles such as carbon black or graphite as a catalyst carrier and a mixture containing a polymer electrolyte as a principal component, and forming a thin film of this paste.

[0005]

Currently, "Nafion" (trade name; manufactured by DuPont.), which is a perfluorocarbon sulfonic acid polymer, is in general use as the polymer electrolyte. In order to provide hydrogen ion conductivity, it is necessary to humidify the electrolyte.

[0006]

The incoming fuel from an anode side is separated into hydrogen ions and electrons on the catalyst of the electrode, while hydrogen ions and electrons that have passed through the electrolyte react with the oxidant on the catalyst on a cathode side. At this time, electric energy can be obtained.

[0007]

In the case where hydrogen is used as the fuel, the reactions below occur in the respective electrodes.

Anode:  $2H_2 \rightarrow 4H^+ + 4e^-$ 

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Cathode:  $O_2 + 4H^+ + 4e^- \rightarrow 2H_2O$ 

Alternatively, in the case where methanol is used as the fuel, the reactions below occur.

Anode:  $CH_3OH + H_2O \rightarrow CO_2 + 6H^+ + 6e^-$ 

Cathode:  $3/2O_2 + 6H^+ + 6e^- \rightarrow 3H_2O$ 

On the catalyst layer inside the electrode, reactants and products are diffused, and the electrons and the hydrogen ions move. Thus, the size of a three-phase zone, which is a reaction point and serves as a passage of each of the fuel, the electrons and the hydrogen ions, becomes important. [0008]

The area of the three-phase zone is an effective area of the catalyst. As this area becomes larger, a utilization factor of the catalyst increases, leading to a higher cell performance. By including the polymer electrolyte in the catalyst layer as described above, the reaction area increases. [0009]

Conventionally, attempts have been made to provide a layer in which the electrode and the polymer electrolyte are mixed and dispersed at an interface between the electrode and the electrolyte. A conventional technology has suggested a method of applying a dispersed solution of the polymer electrolyte and a mixture of catalyst onto a polymer electrolyte membrane and hot-pressing with an electrode, followed by reduction of the catalyst compound, and a method including the reduction, the application and then the hot-pressing (for example, see Patent document 1 and Patent document 2).

[0010]

Further, there has been a method of forming a porous electrode, spraying the polymer electrolyte solution on the electrode, and then hot-pressing this electrode and the polymer electrolyte membrane (for example, see Patent document 3). There also is a method of mixing powder prepared by coating a surface of polymeric resin with a polymer electrolyte into an electrode (for example, see Patent document 5). Moreover, there is a method of mixing a polymer electrolyte, a catalyst, carbon powder and a fluorocarbon resin and forming a film to be an electrode (for example, see Patent document 6).

[0011]

[Patent document 1]

JP 62(1987)-61118 B

[Patent document 2]

JP 62(1987)-61119 B

[Patent document 3]

JP 2(1990)-48632 B

[Patent document 4]

JP 3(1991)-184266 A

[Patent document 5]

JP 3(1991)-295172 A

[Patent document 6]

JP 5(1993)-36418 A

[0012]

[Problem to be solved by the invention]

However, the above-mentioned conventional catalyst layer uses the polymer electrolyte that is soluble in water and an alcohol solution such as ethanol.

[0013]

When an alcohol such as methanol is used as the fuel, a reaction occurs such that alcohol: water = 1:1. Accordingly, during power generation, the electrolyte elutes into the alcohol solution, so that the three—phase zone decreases, lowering the reaction efficiency, and causing a problem of voltage drop.

Furthermore, the electrolyte elutes into water generated in the cathode during power generation and humidifying water necessary for hydrogen ion conduction, so that the three-phase zone decreases, lowering the voltage.

[0015]

[0014]

In order to solve the conventional problems described above, the object of the present invention is to provide a fuel cell using a thin film electrolyte that does not elute into water or alcohol, thereby achieving an increased area of a three-phase zone in a catalyst layer so as to obtain a long lifetime and high voltage.

[0016]

[Means for solving problem]

In order to achieve the above-mentioned object, a fuel cell according

to the present invention is a fuel cell for generating an electric power by supplying one of electrodes with a fuel and the other with an oxidant,

wherein at least one of the electrodes comprises a thin film electrolyte, a catalyst and an electron-conducting substance,

the thin film electrolyte comprises an organic compound molecule that is chemically bonded to a surface of a substrate, and

the organic compound molecule comprises an ion-conducting functional group serving as an electrolyte.
[0017]

It is preferable that the chemical bond is at least one bond selected from the group consisting of a covalent bond, an ionic bond, a coordinate bond and a metallic bond. It is particularly preferable that the chemical bond is a covalent bond formed by an elimination reaction.

[0018]

It is preferable that the chemical bond is a bond to the surface of the substrate via an oxygen atom.
[0019]

It is preferable that the ion-conducting functional group is at least one functional group selected from the group consisting of a phosphonyl group, a phosphinyl group, a sulfonyl group, a sulfonyl group, a carboxyl group, a sulfonic group, a mercapto group, an ether linkage group, a hydroxyl group, a quaternary ammonium group, an amino group and a phosphate group, and contains an ion.

[0020]

It also is preferable that the substrate is a catalyst particle, an electron-conducting moving particle, or an inorganic compound or an organic compound that is further added to the electrode.

[Mode for carrying out the invention]

[0021]

The present invention relates to a fuel cell. In the fuel cell of the present invention, at least one of its catalyst layers has a thin film electrolyte, a catalyst and an electron-conducting substance. The thin film electrolyte contains an organic compound molecule that is chemically bonded to a surface of a substrate, and the organic compound molecule contains an ion-conducting functional group, which serves as an electrolyte. It is preferable that the chemical bond is a covalent bond, an ionic bond, a

coordinate bond or a metallic bond. In particular, a covalent bond formed by an elimination reaction is preferable because it is the most stable in chemical and physical terms. Here, the elimination reaction refers to dehydrohalogenation, dealcoholization (wherein the alcohol has 1 to 3 carbon atoms) or the like.

[0022]

It is preferable that the ion-conducting functional group is at least one functional group selected from the group consisting of a phosphonyl group, a phosphinyl group, a sulfonyl group, a sulfinyl group, a carboxyl group, a sulfonic group, a mercapto group, an ether linkage group, a hydroxyl group, a quaternary ammonium group, an amino group and a phosphate group, and contains an ion. [0023]

It is preferable that the chemical bond is a bond to the surface of the substrate via an oxygen atom. Besides the oxygen atom, the chemical bond may be a bond via a nitrogen atom. This is because the above-mentioned elimination reaction occurs between, for example, a chloro group or an alkoxyl group at the end of a molecule of an organic compound and an active hydrogen, for example, a hydroxyl group (-OH), a carboxyl group (-COOH), an amino group (-NH<sub>2</sub>), or an imino group (>NH), on the substrate surface. [0024]

It is preferable that the substrate is a catalyst particle, an electron-conducting moving particle, or an inorganic compound or an organic compound that is further added to the electrode.

[0025]

The thin film electrolyte and the catalyst in the fuel cell of the present invention may be present on the electron—conducting substance.

[0026]

The thin film electrolyte in the fuel cell of the present invention may be present on the catalyst, and this catalyst may be present on the electron—conducting substance.

[0027]

The thin film electrolyte in the fuel cell of the present invention may be present on a substrate having no electron conductivity.

[0028]

(Examples)

The following is a specific description of the present invention by way of examples.

[0029]

(Example 1)

First, a surface of a catalyst carrying carbon was treated with a hydroxyl group. 5 g of 50 wt% platinum particles having a mean particle diameter of about 3 nm carried by "Ketjen Black EC" (trade name; manufactured by AKZO Chemie, the Netherlands) having a mean primary particle diameter of 30 nm was weighed and put into a 100 cc flask, and then heated from 55°C to 60°C in nitrogen together with a 50 cc fuming sulfuric acid, followed by stirring for 3 days. This was dropped in dehydrated ether that was kept at 0°C, thus obtaining a solid substance. This was stirred with 50 cc distilled water in nitrogen for 10 hours and filtered to obtain a solid substance, which was subjected to vacuum drying. The obtained solid substance is used for a cathode—side electrode. [0030]

Further, 25% platinum particles having a mean particle diameter of about 3 nm and 25% ruthenium particles having a mean particle diameter of about 3 nm that were carried by "Ketjen Black EC" (trade name; manufactured by AKZO Chemie, the Netherlands) having a mean primary particle diameter of 30 nm were prepared, and the carbon surface was treated with a hydroxyl group by the same operation as above. This is used for an anode—side electrode.

[0031]

A thin film electrolyte was formed on the surface of each catalyst carrying carbon that had been treated, and this was formed into a thin film, thus obtaining a catalyst layer.

[0032]

The following is a description of how to form the thin film electrolyte.

[0033]

1 wt% of trichlorosilane compound: CH<sub>2</sub>=CH<sub>-</sub>(CF<sub>2</sub>)<sub>14</sub>(CH<sub>2</sub>)<sub>2</sub>SiCl<sub>3</sub> containing a vinyl group at its end and a fluorocarbon chain at its middle part, which was a reactant, was dissolved in a nonaqueous solvent in which n hexadecane and chloroform were mixed at 4:1. The catalyst carrying carbon that had been subjected to a surface activation treatment was

immersed in this solution for 2 hours. Dehydrochlorination occurred between a hydroxyl group (-OH) present on the catalyst carrying carbon and a chloro group (-Cl) in the chlorosilane compound, so that a monomolecule of the trichlorosilane compound was bonded onto the carbon via oxygen as shown in the formula (1) below.

[0034]

[Formula 1]

Cl Cl Cl Cl CH<sub>2</sub>=CH\_(CF<sub>2</sub>)<sub>14</sub>(CH<sub>2</sub>)<sub>2</sub>Si\_Cl + HO\_substrate 
$$\rightarrow$$
 CH<sub>2</sub>=CH\_(CF<sub>2</sub>)<sub>14</sub>(CH<sub>2</sub>)<sub>2</sub>Si\_O-substrate + HCl Cl Cl

[0035]

After the resultant particles were washed in chloroform, which was a nonaqueous solvent, so as to remove unreacted substances, they were allowed to react with water in the air. Consequently, the monomolecules were bonded to each other via oxygen so as to form a molecular thin film derived from the trichlorosilane compound as shown in the formula (2) below.

[0036]

[Formula 2]

[0037]

Next, the catalyst carrying carbon whose surface was provided with the thin film was allowed to react with a fuming sulfuric acid, whereby an unsaturated bond (a vinyl bond) at the end of the molecule was sulfonated, so that a molecular thin film shown in the formula (3) below was formed. This molecular thin film had a molecular weight of about 912 and a molecular length of 2.8 nm. Here, a SO<sub>3</sub>- group was a group having an ion conductivity, which was formed uniformly on the surface of the molecular thin film in the present embodiment.

[0038]

[Formula 3]

[0039]

This surface—modified catalyst carrying carbon was mixed with water so as to prepare a paste, which was applied to an electrolyte using a thin film electrolyte. Thereafter, this was sandwiched by electrically conductive carbon particle papers treated to be water repellent, which serves as a current collector and hot—pressed, thereby producing a membrane electrode assembly (MEA). FIG. 1 is a drawing showing a structure of the MEA, with numeral 11 denoting the thin film electrolyte and numeral 12 denoting a catalyst layer.

[0040]

The electrolyte part used in the present example was prepared using a thin film electrolyte, whose production method was as follows.

[0041]

A trialkoxysilane compound H+SO<sub>3</sub>-(CH<sub>2</sub>)<sub>2</sub>(CF<sub>2</sub>)<sub>14</sub>(CH<sub>2</sub>)<sub>2</sub>Si(OCH<sub>3</sub>)<sub>3</sub> was pressed into pores of a 100 µm thick porous ceramics, which was an inorganic porous body, having pores with a size on the order of namometers. A demethanolization reaction occurred between a hydroxyl group (-OH) on the ceramics surface and an alkoxy group (in this case, a methoxy group: -OCH<sub>3</sub>), so that a trialkoxysilane compound was bonded to the ceramics as shown in the formula (4) below.

[0042]

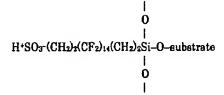
[Formula 4]

[0043]

The monomolecules were bonded to each other by the demethanolization reaction, so that the thin film electrolyte was formed in the pores as shown in the formula (5) below.

[0044]

[Formula 5]



[0045]

Although the molecular film having a sulfonic acid group at its end has been illustrated in the present embodiment, it also is possible to apply not only the sulfonic acid group but also a phosphonyl group, a phosphinyl group, a sulfonyl group, a sulfonyl group, a sulfonic acid group, a mercapto group, a carboxyl group, an ether linkage group, a hydroxyl group, a quaternary ammonium group, an amino group, an ester group and the like as long as it is a functional group providing an ion conductivity. In addition, although a molecular structure having one sulfonic acid group at the end of the molecule has been illustrated in the present example, a molecular structure having a plurality of sulfonic acid groups may be applied as long as the ion conductivity is not lowered.

[0046]

Furthermore, although the catalyst layer is formed of the platinum carrying carbon or the platinum ruthenium carrying carbon and the thin film electrolyte, it also is possible to apply a precious metal catalyst other than platinum and platinum ruthenium, an alloy catalyst, an oxide catalyst or a composite catalyst thereof as the catalyst. Also, the catalyst layer also may contain other additives, for example, a binding agent such as polytetrafluoroethylene (PTFE), a tetrafluoroethylene—hexafluoropropylene copolymer (FEP), polyvinylidene fluoride (PVDF), carbon black or activated carbon, a pore—forming agent such as camphor, CaCO<sub>3</sub> or NH<sub>4</sub>HCO<sub>3</sub>, or a surfactant.

[0047]

(Example 2)

50 wt% platinum having a mean particle diameter of 3 nm carried by "Ketjen Black EC" (trade name: manufactured by AKZO Chemie, the Netherlands) having a mean primary particle diameter of 30 nm was used as a cathode catalyst, while 25 wt% platinum having a mean particle diameter of 3 nm and 25 wt% ruthenium having a mean particle diameter of 3 nm that were carried by the same "Ketjen Black EC" were used as an anode catalyst.

[0048]

This carbon carrying the catalysts, a PTFE dispersion and water were mixed, filtered, and then formed into a sheet by roller pressing. Silica sol having a particle diameter of 80 nm was applied thereto and dried, followed by burning in an argon gas atmosphere at 500°C. [0049]

After burning, a trialkoxysilane compound H+SO<sub>3</sub>-(CH<sub>2</sub>)<sub>2</sub>(CF<sub>2</sub>)<sub>14</sub>(CH<sub>2</sub>)<sub>2</sub>Si(OCH<sub>3</sub>)<sub>3</sub> was pressed into the surface to which the silica sol had been applied. Demethanolization occurred between a hydroxyl group (-OH) on the silica sol surface and an alkoxy group (in this case, a methoxy group: -OCH<sub>3</sub>), so that the trialkoxysilane compound was bonded onto the ceramics. These monomolecules were bonded to each other, thus forming a thin film electrolyte in the pores. [0050]

The trialkoxysilane was filled in not only the silica sol portion but also a solidified catalyst portion, making it possible to produce the catalyst layer and the electrolyte part at one time. Electrically conductive carbon particle papers treated to be water repellent were attached to both external sides of this catalyst layer, and formed in one piece by hot-pressing. [0051]

(Example 3)

The trichlorosilane compound described in Example 1 was bonded to platinum black (oxide) and platinum ruthenium black (oxide) that were in a particulate form according to the method described in Example 1, followed by sulfonation, thus producing a thin film electrolyte. Hydroxyl groups (-OH) on the oxide surfaces in the platinum black and platinum ruthenium black and a chloro group (-Cl) in the trichlorosilane compound reacted with each other, so that a molecular thin film was formed. In other words, since many hydroxyl groups (-OH) are present on the oxide surfaces, the chloro

group (-Cl) reacts in a high-density manner. Next, sulfonation was conducted, thereby forming a thin film electrolyte. The one using the platinum black served as a cathode side catalyst, and the one using the platinum ruthenium black served as an anode side catalyst. [0052]

This surface-modified catalyst and carbon black (acetylene black) were mixed and pressed, thus producing a catalyst layer.
[0053]

The anode side catalyst layer and the cathode side catalyst layer were superposed on each side of the electrolyte part in the thin film electrolyte described in Example 1, and pressed into one piece.

[0054]

(Example 4)

The trichlorosilane compound described in Example 1 was bonded to the surface of silica particles with a diameter of 100 nm (error of ±15 nm) according to the method described in Example 1, followed by sulfonation, thus producing a thin film electrolyte. This was mixed with 50 wt% platinum particles having a mean particle diameter of about 3 nm carried by "Ketjen Black EC" (trade name; manufactured by AKZO Chemie, the Netherlands) having a mean primary particle diameter of 30 nm, or 25% platinum particles having a mean particle diameter of about 3 nm and 25% ruthenium particles having a mean particle diameter of about 3 nm that were carried by the same "Ketjen Black EC" and pressed so as to form a cathode catalyst layer and an anode catalyst layer, respectively. [0055]

A thermal-polymeric organic compound, for example, 3-glycidoxypropyltrimethoxysilane was applied to the thin film electrolyte described in Example 1 that had been produced separately. The above-described catalyst layer was attached to the application surface, while an electrically conductive carbon particle paper treated to be water repellent was attached to the non-application surface, and formed in one piece by hot-pressing. [0056]

As the substrate of the thin film electrolyte, silica spheres were used in the present example. However, other than these, it also is effective to use an inorganic material such as alumina, zeolite, an inorganic oxide or a metallic oxide or a carbon material such as fullerenol, whose surface has or is provided with active hydrogen, a resin material of polyimide, polyurethane, a phenolic resin, polyethylene or polytetrafluoroethylene whose surface has or is provided with active hydrogen, or a composite material thereof.

[0057]

(Example 5)

Fuel cells using the electrode parts illustrated in Examples 1 to 4 will be described as an example.

[0058]

The MEAs obtained by Examples 1 to 4 are expressed by an MEA 1, an MEA 2, an MEA 3 and an MEA 4, respectively. Further, as Comparative Example, an MEA using a polymer electrolyte represented by Nafion as the electrolyte inside the electrode is expressed by an MEA 5. [0059]

How to produce the MEA 1 to MEA 4 is as described in Examples 1 to 4. Also, the MEA 5 was produced as follows.

[0060]

The catalyst layer used here was platinum or platinum ruthenium carried by Ketjen Black EC. 50 wt% platinum having a mean particle diameter of 3 nm carried by "Ketjen Black EC" (trade name; manufactured by AKZO Chemie, the Netherlands) having a mean primary particle diameter of 30 nm was used for a cathode catalyst, while 25 wt% platinum having a mean particle diameter of 3 nm and 25 wt% ruthenium having a mean particle diameter of 3 nm that were carried by the same "Ketjen Black EC" were used for an anode catalyst.

These catalyst carrying particles and a polymer electrolyte were mixed to form a catalyst paste. In this case, the weight of carbon in the catalyst carrying particles to the weight of polymer electrolyte was 1:1. The polymer electrolyte used here was an ethanol isopropanol mixed solution of trade name Flemion (manufactured by ASAHI GLASS CO., LTD.), which was a perfluorocarbon sulfonic acid polymer. [0062]

Next, the above-described catalyst paste was printed on an electrolyte that had been produced by filling a thin film electrolyte in pores

of porous ceramics described in Example 1. A gas diffusion layer on the anode side and a gas diffusion layer on the cathode side were superposed with the electrolyte being at the center, and joined by hot-pressing, thus producing an MEA. As the gas diffusion layers, electrically conductive carbon particle paper (TGPH060H, manufactured by Toray Industries. Inc.) was used.

[0063]

The area of the electrodes of the MEA 1 to the MEA 5 was 5 cm  $\times$  5 cm (length  $\times$  width).

[0064]

In the MEA 1 to the MEA 5 produced as described above, gasket plates made of silicone rubber (with a thickness of 150  $\mu$ m) were joined to an outer peripheral portion of the electrode, and manifolds for cooling water, fuel and oxidant flows were formed. The outer dimension of the gasket was 8 cm  $\times$  8 cm (length  $\times$  width), which was the same as the outer dimension of the unit cell.

[0065]

Subsequently, a separator formed of a 13 mm thick resin-impregnated graphite plate having an outer dimension of 8 cm × 8 cm (length × width) and fuel, oxidant and cooling water channels of 5 mm were prepared. Two separators were used so that the separator provided with the oxidant channel was superposed on one surface of the MEA joined to the gasket plate and the separator provided with the fuel channel was superposed on the other surface thereof, thus forming a unit fuel cell (FIG. 2). In FIG. 2, numeral 21 denotes the membrane electrode assembly (MEA), numeral 22 denotes the gasket plate, numerals 23, 24 and 25 denote the manifolds, and numeral 26 denotes the separator.

The fuel and cooling water to the separator 26 were sent to each cell through the manifolds 23, 24 and 25, flowed through the channels on the separator 26 and were supplied to the MEA.

[0067]

FIG. 3 is a sectional view showing how the unit cells obtained in FIG. 2 were stacked and connected in series. After unit cells 31 and 32 were stacked, they were sandwiched by separators 33 and 34 provided with cooling water channels 43 and 44, and then unit cells 35 and 36 were

connected thereto. By repeating this, an 8-cell-layered cell stack was formed. In other words, adjacent cells were connected in series via the separators 33 and 34. [0068]

In this case, both ends of the cell stack were fixed using stainless steel current collector plates with gold-plated surfaces, insulator plates formed of an electrically insulating material and further end plates and fastening rods. The fastening pressure was 15 kgf/cm<sup>2</sup>. In FIG. 3, numerals 37 to 40 each denote an MEA, numeral 41 denotes a methanol supply port, and numeral 42 denotes an air supply port. [0069]

In the manner described above, respective stacks were layered so as to obtain stacks 1 to 5. With the thus produced stacks 1 to 5, an electric power was generated using a methanol aqueous solution as a fuel. In each stack, 2 mol/L methanol aqueous solution at 60°C was supplied at 2 cc per minute, and the air was humidified by a 60°C bubbler under the condition of a cell temperature of 60°C and an air utilization factor of 30%, thus making measurements. The air outlet was pressurized at 2 atm.

OCV and voltages at 200 mA/cm and 500 mA/cm<sup>2</sup> were as shown in Table 1.
[0071]

[Table 1]

	OCV	200 mA/cm <sup>2</sup>	500 mA/cm <sup>2</sup>
Stack 1	802	522	388
Stack 2	798	512	378
Stack 3	782	501	367
Stack 4	784	505	370
Stack 5	752	426	154

(unit: mV)

[0072]

Even in the case of using 10 mol/L methanol aqueous solution, which was a high concentration methanol aqueous solution, the OCV and the voltages at 200 mA/cm<sup>2</sup> as shown in Table 2 were obtained.

[0073]

[Table 2]

OCV 200 mA/cm<sup>2</sup> Stack 1 401 216 398 212 Stack 2 392 209 Stack 3 204 Stack 4 396 287 89 Stack 5

(unit: mV)

[0074]

Even when using the high concentration methanol solution, the polymer electrolyte monomolecular film did not elute very much. Accordingly, an effective reaction area formed of the catalyst, the fuel and the hydrogen ion conductor, increased, and thus, the voltage rose. [0075]

Incidentally, although methanol was used as an example of the fuel in the present example, the similar result was obtained also by using a hydrocarbon fuel such as hydrogen, ethanol, ethylene glycol, dimethyl ether, isopropanol, ethylene glycol, glycerin, methane or dimethoxymethane, and a mixture thereof. Further, the liquid fuels also may be evaporated in advance and supplied in the form of vapor.

[0076]

Moreover, the structure of the gas diffusion layer of the present example is not limited to the electrically conductive carbon particle paper illustrated in the above examples. Instead, other electrically conductive carbon particle black, electrically conductive carbon particle cloths and metal meshes also can be used effectively.

[0077]

[Effects of the invention]

In accordance with the present invention, at least one of the electrodes has a thin film electrolyte, a catalyst and an electron—conducting substance, so that the elution of the electrolyte in an electrode part catalyst layer and the resulting voltage drop can be suppressed.

## [BRIEF DESCRIPTION OF THE DRAWINGS]

[FIG. 1] FIG. 1 is a schematic plan view showing an electrolyte membrane electrode assembly (MEA) of a fuel cell according to Example 1 of the present invention.

[FIG. 2] FIG. 2 is a perspective view showing a structure of a unit cell of a fuel cell according to Example 5 of the present invention.

[FIG. 3] FIG. 3 is a sectional view showing a stacked structure in which the unit cells of the fuel cell according to Example 5 of the present invention are stacked.

[Explanation of letters or numerals]

- 11 Thin Film Electrolyte
- 12 Catalyst Layer
- 21, 37, 38, 39, 40 Electrolyte Membrane Electrode Assembly (MEA)
- 22 Gasket Plate
- 23, 24, 25 Manifolds
- 26, 33, 34 Separator
- 31, 32, 35, 36 Unit Cell
- 41 Methanol Supply Port
- 42 Air Supply Port
- 43, 44 Cooling Water Channel

[Document Name] ABSTRACT [Abstract]

[Object] A fuel cell using a thin film electrolyte that does not elute into alcohol or water, thereby achieving an increased area of a three—phase zone in a catalyst layer so as to obtain a long lifetime and high voltage is provided.

[Means to Solve the Problems] In a fuel cell for generating an electric power by supplying one (41) of electrodes (37) with a fuel such as methanol and the other (42) with an oxidant such as the air, at least one of the electrodes (37) contains a thin film electrolyte, a catalyst and an electron-conducting substance, the thin film electrolyte contains an organic compound molecule that is chemically bonded to a surface of a substrate, and the organic compound molecule contains an ion-conducting functional group serving as an electrolyte. It is preferable that the chemical bond is at least one bond selected from the group consisting of a covalent bond, an ionic bond, a coordinate bond and a metallic bond. It is preferable that the ion-conducting functional group is a phosphonyl group or the like. [Selected Figure] FIG. 3